Necessity of Time-Reversal Symmetry Breaking for the Polar Kerr Effect in Linear Response

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The Necessity of Time-Reversal Symmetry Breaking for the Polar Kerr Effect in Linear Response

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We show that, measured in a backscattering geometry, the polar Kerr effect is absent if the nonlocal electromagnetic response function respects Onsager symmetry, characteristic of thermodynamic states that preserve time-reversal symmetry. A key element is an expression for the reflectivity tensor in terms of the causal Green's function.

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The polar Kerr effect [1] refers to rotation of the polarization of light upon reflection due to a component of the magnetization perpendicular to the reflecting surface. More generally, it is interpreted as a measure of the corresponding pattern of broken symmetry. Inevitably, a sharp onset of Kerr rotation indicates a symmetry-breaking phase transition. Observation of this phenomenon has indeed played a crucial role in identifying broken time-reversal symmetry in unconventional superconducting states, e.g., of Sr_2RuO_4 [2], UPt_3 [3], and URu_2Si_2 [4]. Kerr onsets have also been found in the pseudogap regime of the cuprates [5–8], although they are somewhat rounded and their interpretation less clear.

In Refs. [2–8], the polar Kerr effect was measured in a backscattering geometry. In the linear response regime, the corresponding Kerr angle is proportional to the ac Hall conductivity in the dimensions perpendicular to the wavevector of light [9, 10] provided that spatial dispersion (nonlocality) is negligible [11]. For this Hall conductivity to be nonzero, the medium must break time-reversal symmetry and mirror symmetries about all planes parallel to the wavevector [12].

However, given the extremely high sensitivity of the Kerr measurements ($\lesssim 100\,\mathrm{nrad}$), it is conceivable that a detectable Kerr rotation arises entirely due to spatial dispersion effects in a medium with zero Hall conductivity. An important question is whether the symmetry requirements discussed in the previous paragraph carry over even if spatial dispersion is taken into account. It is easy to see that the mirror symmetries must be broken in any case. The sense of Kerr rotation is reversed in a mirror parallel to the wavevector, so if the system is invariant under this mirror reflection, the Kerr angle must vanish. Consequences of time-reversal symmetry are more subtle. Kerr rotation is typically described by the macroscopic Maxwell equations which are not invariant under time reversal in the presence of dissipation, even if there is no broken time-reversal symmetry per se.

Indeed, there have been proposals [13, 14] that the polar Kerr effect in backscattering can result from natural optical activity [15], a spatial dispersion effect, even if time-reversal symmetry is unbroken. In a medium with natural optical activity, the speed and damping of circularly polarized light depend on the handedness, and it is plausible that this gives rise to nonzero Kerr rotation. In Refs. [13, 14], the authors computed the Kerr angle in the long-wavelength limit, where spatial dispersion is treated to first order in the wavevector, and obtained nonzero results. Several researchers [16–19] recently adopted this idea to interpret Kerr signals from unconventional superconductors.

Yet there were earlier studies suggesting the contrary [20, 21]. They noted that the macroscopic Maxwell equations should be consistent with the Onsager symmetry [22] of the electromagnetic response function. This is a consequence of *time-reversal symmetry* and *thermal equilibrium*, and holds whether the medium is dissipative or not. In the long-wavelength limit, this consideration leads to electromagnetic boundary conditions (at the boundary of the reflecting medium) different from the ones used in the studies that found nonzero Kerr rotation [13, 14]. The Kerr angle computed with these modified boundary conditions vanishes. Moreover, there were arguments that did not rely on the long-wavelength approximation but reached the same conclusion [23, 24].

The significance of Onsager symmetry has been reemphasized in a number of recent papers [25–28]. This has led to the retraction [29–31] of the proposals that various measured Kerr signals are due to optical activity alone.

Still, as far as we know, the consequences of Onsager symmetry have not been fully clarified. One should be able to see how it constrains the reflectivity tensor in such a way that Kerr rotation is forbidden. However, existing results, if not relying on the long-wavelength limit, either bypass dealing with the reflectivity tensor [23, 26] or take the principle of optical reciprocity [32, 33] as the fundamental postulate [24, 25], but this principle really should be derived from the Onsager symmetry of the response functions. The main obstacle is that without an approximation, there is no obvious way to express the reflectivity tensor in terms of the (nonlocal) response function of the scattering medium. In this work, we avoid this difficulty by expressing the reflectivity tensor in terms of the retarded Green's function of the electromagnetic wave equation. Then, it can be shown that the symmetry of the response function is inherited by the Green's function and hence by the reflectivity tensor. From this we demonstrate, in the framework of nonlocal electrodynamics, that Onsager symmetry leads to optical reciprocity in reflection [34] and, as a special case, the absence of the polar Kerr effect.

We begin by considering the problem of light reflection from an arbitrary linear scattering medium. Without loss of generality, we assume that the medium is entirely in the right half-space ($z > 0$), as in the example illustrated by Fig. 1. Suppose that a right-moving plane wave of frequency ω and transverse wavevector $\mathbf{k}'_\parallel \equiv (k'_x, k'_y)$ is incident on the medium. This wave is of the form $e^{i\mathbf{k}'_+\cdot\mathbf{r}}\,\boldsymbol{\mathcal{E}}_\perp$. (A harmonic time dependence $e^{-i\omega t}$ is assumed throughout.) Here, $\mathbf{k}'_+ \equiv (\mathbf{k}'_\parallel, +k'_z)$ with [35]

$$k'_z \equiv \begin{cases} \operatorname{sgn}(\omega)\sqrt{\omega^2/c^2 - {\mathbf{k}'_\parallel}^2} & (|\mathbf{k}'_\parallel| < |\omega|/c), \\ i\sqrt{{\mathbf{k}'_\parallel}^2 - \omega^2/c^2} & (|\mathbf{k}'_\parallel| \geq |\omega|/c), \end{cases} \tag{1}$$

where c is the speed of light in vacuum; $\boldsymbol{\mathcal{E}}_\perp$ can be any vector satisfying $\mathbf{k}'_+ \cdot \boldsymbol{\mathcal{E}}_\perp = 0$. One should seek for a solution of the electromagnetic wave equation with the following properties: for $z < 0$, the solution is the sum of the incident wave $e^{i\mathbf{k}'_+\cdot\mathbf{r}}\,\boldsymbol{\mathcal{E}}_\perp$ and a left-moving term corresponding to the reflected wave; for $z > 0$, it represents the transmitted wave.

The wave equation is linear, and so is the relationship between the incident and reflected waves. The general form of a reflected (left-moving) wave linearly related to $e^{i\mathbf{k}'_+\cdot\mathbf{r}}\,\boldsymbol{\mathcal{E}}_\perp$ is

$$\int \frac{d^2\mathbf{k}_\parallel}{(2\pi)^2}\, e^{i\mathbf{k}_-\cdot\mathbf{r}}\, \overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel, \mathbf{k}'_\parallel)\cdot \boldsymbol{\mathcal{E}}_\perp \qquad (z<0). \tag{2}$$

Here, $\mathbf{k}_- \equiv (\mathbf{k}_\parallel, -k_z)$, and k_z is defined analogously to k'_z. The *reflectivity tensor* $\overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel, \mathbf{k}'_\parallel)$ satisfies the transversality conditions $\mathbf{k}_- \cdot \overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel, \mathbf{k}'_\parallel) = \overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel, \mathbf{k}'_\parallel)\cdot\mathbf{k}'_+ = 0$.

The backscattered component of the reflected wave is given by the integrand of Eq. (2) at $\mathbf{k}_\parallel = -\mathbf{k}'_\parallel$. Its polarization can be rotated relative to that of the incident wave only if the antisymmetric part of $\overleftrightarrow{\mathbf{R}}_\omega(-\mathbf{k}'_\parallel, \mathbf{k}'_\parallel)$ does not vanish. That is, Kerr rotation is absent when

$$\overleftrightarrow{\mathbf{R}}_\omega(-\mathbf{k}'_\parallel, \mathbf{k}'_\parallel) = \left[\overleftrightarrow{\mathbf{R}}_\omega(-\mathbf{k}'_\parallel, \mathbf{k}'_\parallel)\right]^T. \tag{3}$$

If the medium is uniform in x and y, the reflectivity tensor has the form

$$\overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel, \mathbf{k}'_\parallel) = (2\pi)^2\delta^{(2)}(\mathbf{k}_\parallel - \mathbf{k}'_\parallel)\,\overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}'_\parallel). \tag{4}$$

In this case, Eq. (3) reduces to a simpler condition $\overleftrightarrow{\mathbf{R}}_\omega(0) = \left[\overleftrightarrow{\mathbf{R}}_\omega(0)\right]^T$.

The next step is to relate light reflection to the retarded Green's function in the frequency domain, denoted by $\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r}, \mathbf{r}')$. It is a tensor such that $\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r}, \mathbf{r}')\cdot\hat{\boldsymbol{\xi}}$ is the electric field at $\mathbf{r}$ due to a harmonic point source $i\omega\mu_0\mathbf{J}(\mathbf{r}) = \delta^{(3)}(\mathbf{r}-\mathbf{r}')\hat{\boldsymbol{\xi}}$, where $\hat{\boldsymbol{\xi}}$ is an arbitrary unit vector. In the presence of a medium, $\overleftrightarrow{\mathbf{G}}_\omega$ also includes the effect of scattering. One can decompose the electric field into the incident wave $\overleftrightarrow{\mathbf{G}}^0_\omega(\mathbf{r}, \mathbf{r}')\cdot\hat{\boldsymbol{\xi}}$ and the reflected wave $\left[\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r}, \mathbf{r}') - \overleftrightarrow{\mathbf{G}}^0_\omega(\mathbf{r}, \mathbf{r}')\right]\cdot\hat{\boldsymbol{\xi}}$, where $\overleftrightarrow{\mathbf{G}}^0_\omega$ is the retarded Green's function for free space, provided that both $\mathbf{r}$ and $\mathbf{r}'$ are in the half-space not containing the medium ($z, z' < 0$). This observation enables us to describe light reflection using the Green's function. However, the incident wave here, generated by a point source, is a spherical wave. To make a connection to the reflectivity tensor, it is essential to construct an object that produces an incident plane wave.

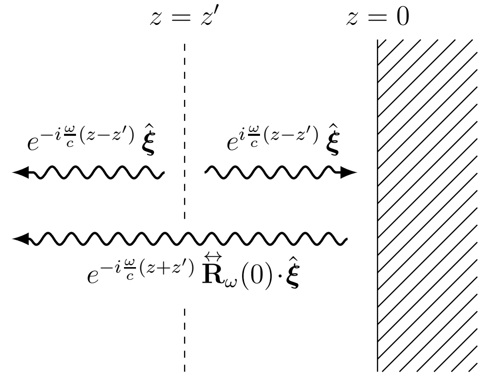

FIG. 1. An example illustrating the relation between the Green's function and the reflectivity tensor: The source $\mathbf{J}(\mathbf{r}) \propto \delta(z-z')\,\hat{\boldsymbol{\xi}}$, where $\hat{\boldsymbol{\xi}} \perp \hat{\mathbf{z}}$, generates a pair of counter-propagating plane waves. One of them is reflected from the medium.

Such an object is in fact well known. Consider the following representation of $\overleftrightarrow{\mathbf{G}}^0_\omega$ [36]:

$$\overleftrightarrow{\mathbf{G}}^0_\omega(\mathbf{r}, \mathbf{r}') = \int \frac{d^2\mathbf{k}'_\parallel}{(2\pi)^2}\, e^{i\mathbf{k}'_\parallel\cdot(\mathbf{r}_\parallel - \mathbf{r}'_\parallel)}\, \overleftrightarrow{\boldsymbol{\mathcal{G}}}^0_\omega(z, z'; \mathbf{k}'_\parallel) \tag{5}$$

with

$$\overleftrightarrow{\boldsymbol{\mathcal{G}}}^0_\omega(z, z'; \mathbf{k}'_\parallel) = \frac{i e^{ik'_z|z-z'|}}{2k'_z}\,\overleftrightarrow{\boldsymbol{\mathcal{P}}}(\mathbf{k}'_\pm) - \frac{c^2}{\omega^2}\delta(z-z')\,\hat{\mathbf{z}}\hat{\mathbf{z}}. \tag{6}$$

Here, $+$ and $-$ are taken for $z > z'$ and $z < z'$, and $\overleftrightarrow{\boldsymbol{\mathcal{P}}}(\mathbf{k}) \equiv \overleftrightarrow{\mathbf{I}} - k^{-2}\mathbf{k}\mathbf{k}$. Thus, the field generated by the planar source distribution $i\omega\mu_0\mathbf{J}(\mathbf{r}) = e^{i\mathbf{k}'_\parallel\cdot\mathbf{r}_\parallel}\delta(z-z')\hat{\boldsymbol{\xi}}$ is

$$\int d^2\mathbf{r}'_\parallel\, e^{i\mathbf{k}'_\parallel\cdot\mathbf{r}'_\parallel}\,\overleftrightarrow{\mathbf{G}}^0_\omega(\mathbf{r}, \mathbf{r}')\cdot\hat{\boldsymbol{\xi}} = e^{i\mathbf{k}'_\parallel\cdot\mathbf{r}_\parallel}\,\overleftrightarrow{\boldsymbol{\mathcal{G}}}^0_\omega(z, z'; \mathbf{k}'_\parallel)\cdot\hat{\boldsymbol{\xi}}. \tag{7}$$

This corresponds to a pair of plane waves with the wavevectors $\mathbf{k}'_\pm$ propagating in the two half-spaces $z > z'$ and $z < z'$.

Now suppose that a scattering medium is placed in one of the half-spaces. We assume that $z' < 0$ and, as before, that the medium is in the region $z > 0$ (e.g., as in Fig.

1). Among the two plane waves, only the one in the half-space $z > z'$ is reflected from the medium. For $z' < z < 0$, the right-hand side of Eq. (7) reduces to $e^{i\mathbf{k}'_+\cdot\mathbf{r}}\boldsymbol{\mathcal{E}}_\perp$ with $\boldsymbol{\mathcal{E}}_\perp = i(2k'_z)^{-1}e^{-ik'_z z'}\overleftrightarrow{\boldsymbol{\mathcal{P}}}(\mathbf{k}'_+)\cdot\hat{\boldsymbol{\xi}}$. The resulting reflected wave can be either obtained from Eq. (2) or expressed in terms of the Green's function. Thus, for $z, z' < 0$,

$$\begin{aligned}&\frac{ie^{-ik'_z z'}}{2k'_z}\int\frac{d^2\mathbf{k}_\parallel}{(2\pi)^2}\,e^{i\mathbf{k}_-\cdot\mathbf{r}}\,\overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\cdot\overleftrightarrow{\boldsymbol{\mathcal{P}}}(\mathbf{k}'_+)\cdot\hat{\boldsymbol{\xi}}\\&=\int d^2\mathbf{r}'_\parallel\,e^{i\mathbf{k}'_\parallel\cdot\mathbf{r}'_\parallel}\big[\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r},\mathbf{r}')-\overleftrightarrow{\mathbf{G}}^0_\omega(\mathbf{r},\mathbf{r}')\big]\cdot\hat{\boldsymbol{\xi}},\end{aligned}\tag{8}$$

and $\overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\cdot\overleftrightarrow{\boldsymbol{\mathcal{P}}}(\mathbf{k}'_+) = \overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$ due to transversality. As $\hat{\boldsymbol{\xi}}$ is arbitrary, Eq. (8) without the contraction with $\hat{\boldsymbol{\xi}}$ holds as a tensor identity. Then, the Fourier transform with respect to $\mathbf{r}_\parallel$ allows us to express the reflectivity tensor in terms of the Green's function as

$$\begin{aligned}&\frac{i}{2k'_z}e^{-i(k_z z+k'_z z')}\,\overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\\&=\overleftrightarrow{\boldsymbol{\mathcal{G}}}_\omega(z,z';\mathbf{k}_\parallel,\mathbf{k}'_\parallel)-\overleftrightarrow{\boldsymbol{\mathcal{G}}}^0_\omega(z,z';\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\quad(z,z'<0),\end{aligned}\tag{9}$$

where

$$\overleftrightarrow{\boldsymbol{\mathcal{G}}}_\omega(z,z';\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\equiv\int d^2\mathbf{r}_\parallel d^2\mathbf{r}'_\parallel\,e^{-i(\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel-\mathbf{k}'_\parallel\cdot\mathbf{r}'_\parallel)}\,\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r},\mathbf{r}')\tag{10}$$

and

$$\overleftrightarrow{\boldsymbol{\mathcal{G}}}^0_\omega(z,z';\mathbf{k}_\parallel,\mathbf{k}'_\parallel)=(2\pi)^2\delta^{(2)}(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)\,\overleftrightarrow{\boldsymbol{\mathcal{G}}}^0_\omega(z,z';\mathbf{k}'_\parallel).\tag{11}$$

As will be shown later, the Onsager symmetry of the electromagnetic response function implies

$$\overleftrightarrow{\boldsymbol{\mathcal{G}}}_\omega(z,z';\mathbf{k}_\parallel,\mathbf{k}'_\parallel)=\big[\overleftrightarrow{\boldsymbol{\mathcal{G}}}_\omega(z',z;-\mathbf{k}'_\parallel,-\mathbf{k}_\parallel)\big]^T.\tag{12}$$

Eqs. (6) and (11) show that $\overleftrightarrow{\boldsymbol{\mathcal{G}}}^0_\omega$ also satisfies the analogous relation. (This can be viewed as a special case of the above equation.) From Eqs. (9) and (12), the reciprocity relation for light reflection follows:

$$\frac{1}{k'_z}\overleftrightarrow{\mathbf{R}}_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)=\frac{1}{k_z}\big[\overleftrightarrow{\mathbf{R}}_\omega(-\mathbf{k}'_\parallel,-\mathbf{k}_\parallel)\big]^T,\tag{13}$$

which was previously proved under the assumption that spatial dispersion effects are negligible [34]. Eq. (13) reduces to Eq. (3) for backscattering ($\mathbf{k}_\parallel = -\mathbf{k}'_\parallel$) and hence implies the absence of Kerr rotation.

As a side note, a similar construction can also be applied to light transmission. If the scattering medium has a finite extent in the z-direction, one can define, in a manner analogous to the reflectivity tensor, the transmissivity tensors for right- and left-moving waves (denoted as $\overleftrightarrow{\mathbf{T}}^\pm_\omega$). It is straightforward to verify that for z and z' outside of and on opposite sides of the medium,

$$\frac{i}{2k'_z}e^{\pm i(k_z z-k'_z z')}\,\overleftrightarrow{\mathbf{T}}^\pm_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)=\overleftrightarrow{\boldsymbol{\mathcal{G}}}_\omega(z,z';\mathbf{k}_\parallel,\mathbf{k}'_\parallel).\tag{14}$$

Here, $+$ and $-$ are taken for $z > z'$ and $z < z'$, and the transversality conditions read $\mathbf{k}_\pm\cdot\overleftrightarrow{\mathbf{T}}^\pm_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \overleftrightarrow{\mathbf{T}}^\pm_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\cdot\mathbf{k}'_\pm = 0$. Eqs. (12) and (14) lead to the reciprocity relation for transmission [34]:

$$\frac{1}{k'_z}\overleftrightarrow{\mathbf{T}}^+_\omega(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)=\frac{1}{k_z}\big[\overleftrightarrow{\mathbf{T}}^-_\omega(-\mathbf{k}'_\parallel,-\mathbf{k}_\parallel)\big]^T.\tag{15}$$

We now derive Eq. (12). Consider general linear constitutive relations for time-harmonic fields in nonlocal electrodynamics:

$$\mathbf{D}=\widetilde{\boldsymbol{\epsilon}}_\omega\mathbf{E},\qquad\mathbf{H}=\mu_0^{-1}\mathbf{B},\tag{16}$$

where the permittivity operator $\widetilde{\boldsymbol{\epsilon}}_\omega$ is defined by

$$\frac{1}{\epsilon_0}\big(\widetilde{\boldsymbol{\epsilon}}_\omega\mathbf{E}\big)(\mathbf{r})\equiv\mathbf{E}(\mathbf{r})+\int d^3\mathbf{r}'\,\overleftrightarrow{\boldsymbol{\chi}}_\omega(\mathbf{r},\mathbf{r}')\cdot\mathbf{E}(\mathbf{r}').\tag{17}$$

We have adopted the Landau-Lifshitz approach (§103 of [37]), in which the effect of the medium is solely contained in $\widetilde{\boldsymbol{\epsilon}}_\omega$, i.e., the permeability is simply taken to be the constant μ_0. The kernel $\overleftrightarrow{\boldsymbol{\chi}}_\omega$ is the electromagnetic response function (nonlocal susceptibility tensor). From the linear response theory, it can be shown that time-reversal symmetry and thermal equilibrium lead to the following Onsager symmetry relation [22]:

$$\overleftrightarrow{\boldsymbol{\chi}}_\omega(\mathbf{r},\mathbf{r}')=\big[\overleftrightarrow{\boldsymbol{\chi}}_\omega(\mathbf{r}',\mathbf{r})\big]^T.\tag{18}$$

The dynamics of the fields is given by the macroscopic Maxwell equations:

$$\begin{aligned}&\nabla\times\mathbf{E}=i\omega\mathbf{B},\qquad&&\nabla\cdot\mathbf{B}=0,\\&\nabla\cdot\mathbf{D}=\rho,&&\nabla\times\mathbf{H}=\mathbf{J}-i\omega\mathbf{D}.\end{aligned}\tag{19}$$

Eqs. (16) and (19) lead to an integro-differential equation for $\mathbf{E}$, the electromagnetic wave equation:

$$\widetilde{\mathcal{L}}_\omega\mathbf{E}\equiv(\omega\mu_0)^{-1}i\triangle_t\mathbf{E}+i\omega\widetilde{\boldsymbol{\epsilon}}_\omega\mathbf{E}=\mathbf{J},\tag{20}$$

where $\triangle_t$ is the transverse Laplacian defined by $\triangle_t\mathbf{E}\equiv-\nabla\times(\nabla\times\mathbf{E})$. Recall that we assume the scattering medium is entirely in the region $z>0$; that is, $\overleftrightarrow{\boldsymbol{\chi}}_\omega(\mathbf{r},\mathbf{r}')=0$ if $z<0$ or $z'<0$ [38]. Hence,

$$\big(\widetilde{\mathcal{L}}_\omega\mathbf{E}\big)(\mathbf{r})=\frac{i}{\omega\mu_0}\left(\triangle_t+\frac{\omega^2}{c^2}\right)\mathbf{E}(\mathbf{r})\quad(z<0).\tag{21}$$

The Green's function is the kernel associated with the operator $(i\omega\mu_0)^{-1}\widetilde{\mathcal{L}}_\omega^{-1}$:

$$\frac{1}{i\omega\mu_0}\big(\widetilde{\mathcal{L}}_\omega^{-1}\mathbf{J}\big)(\mathbf{r})\equiv\int d^3\mathbf{r}'\,\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r},\mathbf{r}')\cdot\mathbf{J}(\mathbf{r}').\tag{22}$$

This definition shows that the symmetry of $\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r},\mathbf{r}')$ is inherited from that of $\widetilde{\mathcal{L}}_\omega$. In particular, $\widetilde{\mathcal{L}}_\omega$ is a complex symmetric operator, i.e.,

$$\langle\mathbf{E}_1,\widetilde{\mathcal{L}}_\omega\mathbf{E}_2\rangle=\langle\widetilde{\mathcal{L}}_\omega\mathbf{E}_1,\mathbf{E}_2\rangle,\tag{23}$$

for $\mathbf{E}_1$ and $\mathbf{E}_2$ belonging to a suitable class of vector-valued functions. Here, we define

$$\langle \mathbf{u}, \mathbf{v} \rangle \equiv \int d^3\mathbf{r}\, \mathbf{u}(\mathbf{r}) \cdot \mathbf{v}(\mathbf{r}). \tag{24}$$

Eq. (23) follows from Eq. (18) and that $\langle \mathbf{E}_1, \triangle_t \mathbf{E}_2 \rangle = \langle \triangle_t \mathbf{E}_1, \mathbf{E}_2 \rangle$ (integration by parts without a surface term). Defining $\mathbf{J}_1 \equiv \widetilde{\mathcal{L}}_\omega \mathbf{E}_1$ and $\mathbf{J}_2 \equiv \widetilde{\mathcal{L}}_\omega \mathbf{E}_2$, we see that $\widetilde{\mathcal{L}}_\omega^{-1}$ is also complex symmetric:

$$\langle \widetilde{\mathcal{L}}_\omega^{-1} \mathbf{J}_1, \mathbf{J}_2 \rangle = \langle \mathbf{J}_1, \widetilde{\mathcal{L}}_\omega^{-1} \mathbf{J}_2 \rangle. \tag{25}$$

Together with Eq. (22), this implies the symmetry relation

$$\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r}, \mathbf{r}') = \left[\overleftrightarrow{\mathbf{G}}_\omega(\mathbf{r}', \mathbf{r})\right]^T, \tag{26}$$

and Eq. (12) follows by virtue of Eq. (10).

In the above analysis, one should ensure that $\overleftrightarrow{\mathbf{G}}_\omega$ or, equivalently, $\widetilde{\mathcal{L}}_\omega^{-1}$ is uniquely determined. Notice that $\overleftrightarrow{\mathbf{G}}_\omega$ is the retarded Green's function. In other words, $\widetilde{\mathcal{L}}_\omega^{-1} \mathbf{J}$ is an outgoing wave (with the effect of scattering included), meaning that no radiation originates from infinity. [Only then can the right-hand side of Eq. (8) be identified as a pure reflected wave.] Intuitively, we expect that restricting $\overleftrightarrow{\mathbf{G}}_\omega$ to be the retarded Green's function guarantees its uniqueness; a given current distribution should result in a unique outgoing electromagnetic field distribution.

It is possible to make the uniqueness argument more formal. We sketch the arguments here; a more thorough mathematical treatment is given in the Supplemental Material [39]. As is often done to ensure causality, we add an arbitrarily small positive imaginary part to the frequency ($\omega \rightarrow \omega_+ \equiv \omega + i\eta$) [40]. The frequency shift $i\eta$ makes the entire space slightly dissipative. This fact is readily seen for a homogeneous, lossless, propagating medium. Consider a plane wave proportional to $e^{i\mathbf{k}\cdot\mathbf{r}}$ and a dispersion relation $\omega = f(\mathbf{k})$. Upon shifting ω by $i\eta$, $\mathbf{k}$ should change by an amount $\delta\mathbf{k}$ satisfying $\delta\mathbf{k} \cdot (\partial f / \partial \mathbf{k}) = i\eta$ to maintain the dispersion relation. The additional factor $e^{i\,\delta\mathbf{k}\cdot\mathbf{r}}$ exponentially decays along the direction of propagation (parallel to $\partial f / \partial \mathbf{k}$).

In the presence of the dissipation introduced by $i\eta$, a wave $\mathbf{E}(\mathbf{r})$ originating from infinitely far away must have a divergent amplitude there. Otherwise it vanishes at any finite $\mathbf{r}$, i.e., after having traveled and been dissipated over an infinite distance. For the same reason, we expect that a wave is exponentially suppressed at infinity if and only if it is outgoing. Two such waves $\mathbf{E}_1$ and $\mathbf{E}_2$ satisfy $\langle \mathbf{E}_1, \triangle_t \mathbf{E}_2 \rangle = \langle \triangle_t \mathbf{E}_1, \mathbf{E}_2 \rangle$ (i.e., no surface term arises) and hence Eq. (23). Moreover, one can select outgoing waves by demanding $\mathbf{E}$ to be square integrable. This requirement, in particular, excludes nontrivial solutions of the homogeneous equation $\widetilde{\mathcal{L}}_{\omega_+} \mathbf{E} = 0$ because they correspond to waves generated at infinity. It then follows that the solution of the inhomogeneous equation $\widetilde{\mathcal{L}}_{\omega_+} \mathbf{E} = \mathbf{J}$ is unique.

In summary, we have shown, in the framework of nonlocal electrodynamics, that the Onsager symmetry of the electromagnetic response function implies the absence of Kerr rotation in backscattering and, more generally, the principle of optical reciprocity. An important observation is that the symmetry property of the response function is inherited by the Green's function and then by the reflectivity tensor.

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[38] Notice that $z = 0$ does not need to be a physical boundary. The medium should be well within the region $z > 0$ so that its optical response has no impact on $z < 0$.

[39] See Supplemental Material, which includes Refs. [41–46].

[40] Instead of using Eq. (1), one can equivalently define k'_z as $\left(\omega_+^2/c^2 - \mathbf{k}'^2_\parallel\right)^{1/2}$, where the branch cut of the complex square root function is taken to be the positive real axis.

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